Eliashberg theory of superconductivity and inelastic rare-earth impurity scattering in filled skutterudite $La_{1-x}Pr_xOs_4Sb_{12}$

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We study the influence of inelastic rare-earth impurity scattering on electron-phonon-mediated superconductivity and mass renormalization in $La_{1-x}Pr_xOs_4Sb_{12}$ compounds. Solving the strong-coupling Eliashberg equations we find that the dominant quadrupolar component of the inelastic scattering on Pr impurities yields an enhancement of the superconducting transition temperature T_c in $LaOs_4Sb_{12}$ and increases monotonically as a function of Pr concentration. The calculated results are in good agreement with the experimentally observed $T_c(x)$ dependence. Our analysis suggests that phonons and quadrupolar excitations cause an attractive electron interaction which results in the formation of Cooper pairs and singlet superconductivity in $PrOs_4Sb_{12}$.

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The Pr-based filled-skutterudite compounds have attracted much attention because of their exotic properties like metal-insulator transition or unusual heavy-fermion behavior.[1,2,3] This also concerns recently discovered $PrOs_4Sb_{12}$, the first Pr-based heavy-fermion superconductor with $T_c = 1.85$ K which possesses many exotic properties compared to that of the Ce- and U-based superconductors.[4] The heavy-electron mass has been confirmed by the large specific heat jump $\Delta C/T_c \sim 500\ mJ/(K^2\ mol)$ at T_c (see Ref. 4) and by de Haas-van Alphen (dHvA) measurements.[5] The ground state in the crystalline electric field (CEF) for Pr^{3+} ion is a Γ_1 singlet, which is separated by the first excited state, a $\Gamma_4^{(2)}$ triplet by a gap of $\Delta_{CEF} \sim 8$ K.[6] Because of the small Δ_{CEF}, the relation between the quadrupole fluctuations associated with the $\Gamma_4^{(2)}$ state and the superconductivity has been recently the focus of intense discussions.[7,8]

At present, the Cooper-pairing mechanism and the corresponding symmetry of the superconducting gap in $PrOs_4Sb_{12}$ is still under debate. For example, initial studies of the thermal conductivity in a rotated magnetic field have suggested the presence of two distinct SC phases.[9] Similarly, the London penetration depth[10] has indicated a possible nodal structure of the superconducting gap. On the contrary, a number of experimental techniques such as nuclear-quadrupole-resonance (NQR),[11,12] Scanning Tunneling Microscopy(STM),[13] muon spin relaxation, (μSR)[14] thermal conductivity,[15] and specific heat measurements[16] are in agreement with a fully developed isotropic s-wave-like superconducting gap at the Fermi surface (FS). Another interesting experimental observation by means of the dHvA effect was that $PrOs_4Sb_{12}$ has a very similar FS topology as the conventional s-wave superconductor $LaOs_4Sb_{12}$ (Ref. 5) ($T_c = 0.74$ K) which hints at a relatively weak hybridization of the conduction band with Pr^{3+} $4f^2$ electrons. Furthermore, it has been found[12,17] that in the alloy compound $La_{1-x}Pr_xOs_4Sb_{12}$ the superconducting transition temperature changes smoothly upon changing x. These observations raise doubts about the unconventional nature of the Cooper pairing in $PrOs_4Sb_{12}$. But the enhancement of T_c and the effective quasiparticle mass in this compound with respect to that of $LaOs_4Sb_{12}$ have to be understood.

It is well known that scattering by magnetic impurities suppresses conventional s-wave superconductivity by destroying Cooper pairs in a singlet state. This situation, however, can change in the case of paramagnetic non-Kramers rare-earth impurities. For example, it has been shown previously[18] that for superconductors containing an impurity with crystal-field-split energy levels the inelastic charge scattering of conduction electrons from the aspherical part of the $4f$ charge distribution may yield an increase of T_c. However, in most of the cases the usual magnetic exchange interaction is dominant, thus suppressing T_c. In this Rapid Communication, we solve the nonlinear Eliashberg equations for electron-phonon mediated Cooper pairing including inelastic scattering on Pr impurities. We find that a dominant quadrupolar scattering introduced by the Pr ions is responsible for an increase of T_c in $La_{1-x}Pr_xOs_4Sb_{12}$ as a function of Pr concentration.

Before solving the nonlinear version of the Eliashberg equations let us illustrate the results of Ref. 18 where the influence of inelastic scattering by rare-earth impurities on the superconducting transition temperature T_{c0} without impurities has been studied for both exchange and quadrupolar scattering. In particular, for impurities with two singlet levels separated by an energy Δ_{CEF} the analog of the Abrikosov-Gor'kov relation for the change of T_c as a function of the total impurity scattering rate[18] for the s-wave superconductor is given by

$$-\frac{8}{\pi}T_c\tau_{12}^M \ln\frac{T_c}{T_{c0}} \;=\; 1+\frac{\tanh x}{x}-(\tanh x)^2-A(x)+B(x),$$

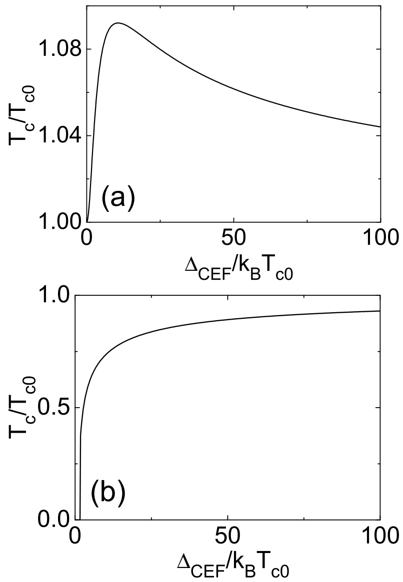

FIG. 1: Calculated values of the superconducting transition temperatures as a function of Δ_{CEF}/T_{c0} for inelastic quadrupolar (a) and magnetic (b) scattering by impurities with two singlet levels using Eqs. (1).

$$-\frac{8}{\pi}T_c\tau_{12}^Q\ln\frac{T_c}{T_{c0}} \;=\; 1-\frac{\tanh x}{x}-(\tanh x)^2+A(x). \qquad (1)$$

Here, $1/\tau_{12}^M$ and $1/\tau_{12}^Q$ are the magnetic and quadrupolar scattering rate, respectively, and $x=\Delta_{CEF}/2k_BT_{c0}$. Furthermore, $A(x)$ and $B(x)$ are the combinations of digamma functions[18] and are smooth functions of x. In Figs. 1(a) and 1(b) we show the behavior of T_c/T_{c0} as a function of Δ_{CEF}/k_BT_{c0} for the quadrupolar (charge) and exchange (magnetic) scattering, respectively. Assuming equal matrix elements for both scatterings ($\tau_{12}^M T_{c0}=\tau_{12}^Q T_{c0}=\tau_{12}T_{c0}$) and setting somewhat arbitrarily $\tau_{12}T_{c0}=1.4$ we find very different behavior. While for quadrupolar scattering T_c increases with respect to T_{c0}, with a pronounced maximum around $\Delta_{CEF}/k_BT_{c0}\approx 10$, the magnetic scattering results in a complete suppression of T_c at small Δ_{CEF} and gives a moderate reduction at larger x values. Overall one finds that for similar scattering amplitudes the magnetic exchange scattering dominates and T_c decreases.

Let us consider more specifically Pr^{3+} impurities in $La_{1-x}Pr_xOs_4Sb_{12}$ compounds. As mentioned above the CEF level scheme is composed of a Γ_1 singlet ground state, a $\Gamma_4^{(2)}$ triplet first excited state with $\Delta_{CEF}\approx 8$ K, and other excited states located at much larger energies above ~ 100 K.[6] Furthermore, due to T_h crystal group symmetry, the $\Gamma_4^{(2)}$ level is of the form $|\Gamma_4^{(2)}(m)\rangle=\sqrt{1-d^2}|\Gamma_5(m)\rangle+d|\Gamma_4(m)\rangle$, where $m=+,-,0$. Here, Γ_5 and Γ_4 are the wave functions for the cubic O_h symmetry and d is a parameter that describes the mixture of the two states and is estimated to be equal 0.26.[19] Since scattering between Γ_1 and Γ_5 is quadrupolar, the dominant scattering between $\Gamma_4^{(2)}$ and Γ_1 will be quadrupolar as well. Furthermore, from Fig. 1(a) we see that the enhancement of T_c is largest for $\Delta_{CEF}\approx 8$ K. Keeping in mind that the higher excited states lie at energies ≥ 100 K the magnetic pair-breaking scattering from these levels will play only a minor role in affecting T_c. This suggests that Pr^{3+}ions in $La_{1-x}Pr_xOs_4Sb_{12}$ yield a strong Cooper-pair enhancement as compared with $LaOs_4Sb_{12}$. At the same time, the Pr^{3+} ions in $La_{1-x}Pr_xOs_4Sb_{12}$ can be treated as independent impurities only at low enough doping. At large Pr concentrations the RKKY interaction between the $4f^2$ ions results in the formation of O_{xy}-type quadrupolar exciton bands as confirmed experimentally.[8] The latter can be treated as low-energy bosonic excitations which contribute in addition to phonons to the Cooper-pairing. But one should notice that the exciton dispersion is relatively weak, *i.e.*, about 10% of Δ_{CEF}.[8]

Using the generalized Holstein-Primakoff (HP) method,[20] the low-energy Hamiltonian describing the magnetic and quadrupolar interaction between conduction electrons and the CEF split energy levels of Pr^{3+} is given by

$$H=\sum_{\mathbf{q}u}\omega_{\mathbf{q}}\beta^\dagger_{\mathbf{q}u}\beta_{\mathbf{q}u}-I_{ac}\sum_{\mathbf{k},\mathbf{q},\sigma,u}f_u(\mathbf{q})\lambda^Q_{\mathbf{q}}\hat{O}_{\mathbf{q}u}c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}+\mathbf{q}\sigma}$$
$$-(1-g_L)I_{ex}\sum_{\mathbf{k},\mathbf{q},\sigma,\sigma',u}\sigma^u_{\sigma\sigma'}\lambda^M_{\mathbf{q}}\hat{J}_{\mathbf{q}u}c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}+\mathbf{q}\sigma'}, \qquad (2)$$

where $\omega_{\mathbf{q}}$ is the energy dispersion of the exciton. There are three bosonic modes ($u=a,b,c$) corresponding to the excitations between singlet Γ_1 and triplet $\Gamma_4^{(2)}$ states. σ^u are the Pauli matrices ($c=x$, $b=z$, $a=y$) and $f_c(\mathbf{q})=\hat{q}_x\hat{q}_y$, $f_b(\mathbf{q})=\hat{q}_z\hat{q}_x$, and $f_a(\mathbf{q})=\hat{q}_y\hat{q}_z$ are the form factors of the quadrupolar interaction. The quadrupolar and magnetic excitations are written in terms of the bosonic operators $\hat{O}_{\mathbf{q}u}=i\left(\beta_{\mathbf{q}u}-\beta^\dagger_{-\mathbf{q}u}\right)$ and $\hat{J}_{\mathbf{q}u}=\left(\beta_{\mathbf{q}u}+\beta^\dagger_{-\mathbf{q}u}\right)$ with $\left(\lambda^Q_{\mathbf{q}}\right)^2=(\Delta_{CEF}+2D_Mz\gamma_{\mathbf{q}})/\omega_{\mathbf{q}}$ and $\left(\lambda^M_{\mathbf{q}}\right)^2=(\Delta_{CEF}+2D_Qz\gamma_{\mathbf{q}})/\omega_{\mathbf{q}}$, respectively. Here, D_M and D_Q are the effective magnetic and quadrupolar coupling constants,[19] z is the coordination number, and $\gamma_{\mathbf{k}}$ is the corresponding structure factor. Due to the weak dispersion of the excitons, $\lambda^Q_{\mathbf{q}}$ and $\lambda^M_{\mathbf{q}}$ are almost momentum independent. Solving the Eliashberg equations, we use an effective interaction between quadrupolar excitons and conduction electrons averaged over the FS.

On the real frequency axis the finite temperature Eliashberg equations for the superconducting gap $\Delta(\omega,T)$ and the renormalization function $Z(\omega,T)$ are given by[21]

$$\Delta(\omega,T) \;=\; \frac{1}{Z(\omega,T)}\int_0^\infty d\omega'\mathrm{Re}\left\{\frac{\Delta(\omega',T)}{\sqrt{\omega'^2-\Delta^2(\omega',T)}}\right\}$$
$$\times\;\left[K_+(\omega,\omega',T)-\mu^*\tanh\left(\frac{\beta\omega'}{2}\right)\right], \qquad (3)$$

$$\omega(1 - Z(\omega, T)) = \int_0^{\infty} d\omega' \mathrm{Re}\left\{\frac{\omega'}{\sqrt{\omega'^2 - \Delta^2(\omega', T)}}\right\} \times K_-(\omega, \omega', T), \quad (4)$$

where

$$K_\pm(\omega, \omega', T) = \int_0^{\infty} d\Omega H_\pm(\Omega) \times \left[\frac{f(-\omega') + n(\Omega)}{\omega' + \omega + \Omega} \pm \frac{f(-\omega') + n(\Omega)}{\omega' - \omega + \Omega} \mp \frac{f(\omega') + n(\Omega)}{-\omega' + \omega + \Omega} - \frac{f(\omega') + n(\Omega)}{-\omega' - \omega + \Omega}\right], \quad (5)$$

and $H_\pm(\Omega) = \alpha_P^2 G(\Omega) + \alpha_Q^2 F(\Omega) \mp \alpha_M^2 F(\Omega)$ is the generalized electron-boson coupling function averaged over the FS. Here, μ^* is the screened Coulomb repulsion, $f(\omega)$ and $n(\omega)$ are the Fermi and Bose distribution functions, respectively, and $\beta = 1/k_B T$.

As mentioned above there are two types of bosonic excitations. In $LaOs_4Sb_{12}$ the superconductivity is driven by the electron-phonon interaction and we assume a single Lorentzian phonon mode at $\hbar\omega_E = 26$ meV with broadening $\Gamma_p = \omega_E/5$. This corresponds to the Debye temperature of this compound[22] (see Fig. 2). Setting

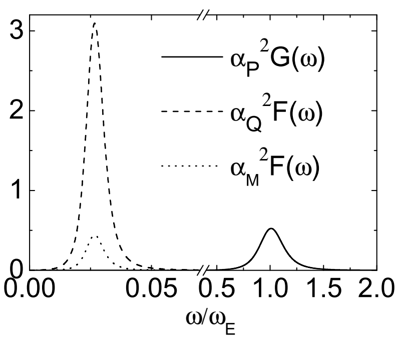

FIG. 2: Calculated bosonic Eliashberg function $H_\pm(\Omega)$ for $La_{1-x}Pr_xOs_4Sb_{12}$. The solid curve denotes the phonon contribution while the dashed and dotted curves refer to the quadrupolar and magnetic contributions, respectively.

$\mu^* = 0.1$ and $\lambda = 2\int_{-\infty}^{+\infty} d\Omega \alpha_P^2 G(\Omega)/\Omega = 0.33$ the Eliashberg equations yield $T_c = 0.74$ K and $2\Delta_0/k_B T_c \approx 3.5$. In Fig. 3(a) we show the calculated results for the real part of $\Delta(\omega)$ and the renormalization function $Z(\omega)$. In both cases one finds typical behavior of a BCS-like superconductor. In particular, $\mathrm{Re}\Delta(\omega)$ shows a peak structure at ω_E and becomes negative at larger frequencies, reflecting the effective repulsion for $\omega > \omega_E$. For $\omega_E >> \Delta_0$, the renormalization function is close to 1 and the ratio $2\Delta_0/k_B T_c$ equals the BCS value.

A similar crystallographic structure and nearly equal ionic radii of the La and Pr ions allow to assume nearly the same phonon modes in $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$. The main difference is the occurrence of the low-energy exciton in $PrOs_4Sb_{12}$ at about $\Delta_{CEF} \approx 0.03\omega_E$. As discussed above there are both (magnetic) pair-breaking and

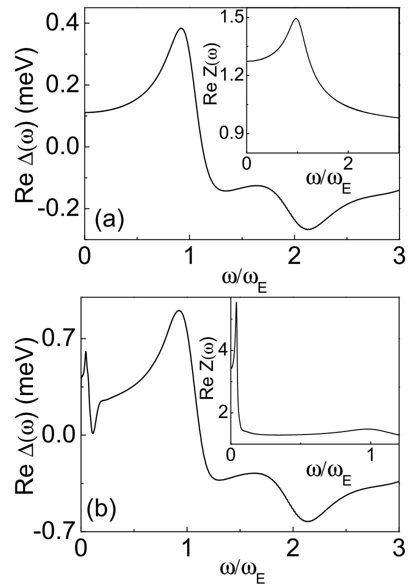

FIG. 3: Calculated frequency dependence of the real part of the superconducting gap function $\Delta(\omega)$ for $LaOs_4Sb_{12}$ (a) and $PrOs_4Sb_{12}$ (b) compounds. The renormalization function, $Z(\omega)$ at $T = 0.04$ K is shown in the insets. Note that we set the cutoff frequency equal to $5\omega_E$ and introduce a finite damping $\Gamma = 0.01$ meV.

(quadrupolar) pair-forming scattering processes of electrons by the excitons. Both processes can be taken into account as separate contributions to the Eliashberg function $\alpha^2 F(\omega)$. Using Eq. (2) and ignoring a weak dispersion of the exciton as observed by inelastic neutron scattering (INS) experiments[8] we obtain, after averaging over the FS, $\lambda_Q = 2\int_0^{\infty} d\omega \alpha_Q^2 F(\omega)/\omega \approx I_{ac}^2 |M_{14}^Q|^2 2N_0/\Delta_{CEF}$ where $|M_{14}^Q|^2 = 35(1 - d^2)$ is the quadrupolar scattering matrix element and $N_0 = 7.625$ eV^{-1} is the bare (unrenormalized) electronic density of states.[22] Furthermore, we set $I_{ac} \approx 2$ meV. Similarly, we find $\lambda_M = 2\int_0^{\infty} d\omega \alpha_M^2 F(\omega)/\omega \approx (1 - g_L)^2 I_{ex}^2 |M_{14}^M|^2 2N_0/\Delta_{CEF}$ where, $M_{14}^M = 20d^2/3$ is the magnetic scattering matrix element and g_L =0.8 is the Lande$'$ factor. Following our previous estimates, we set $\alpha_Q^2/\alpha_M^2 = 7$ and obtain $I_{ex} \approx 16.4 I_{ac}$, where I_{ex} is the on-site magnetic interaction constant between exciton and conduction electrons. All components entering the total Eliashberg function $H_\pm(\Omega)$ are shown in Fig. 2. As expected the quadrupolar contribution is about 7 times larger than the magnetic one. Therefore an inclusion of the excitons yields an enhancement of the superconductivity in $PrOs_4Sb_{12}$ as compared to $LaOs_4Sb_{12}$. In particular, the total value of λ increases to 3.05 yielding strong coupling superconductivity in $PrOs_4Sb_{12}$ with $T_c = 1.85$ K. Here, one has to remember that in $PrOs_4Sb_{12}$ there are three contributions to the total λ and their sum gives λ=3.05. This yields a strong renormalization of the quasiparticle mass in $PrOs_4Sb_{12}$ compared to its $LaOs_4Sb_{12}$ counterpart. At the same time T_c and $\Delta(\omega)$ are determined by Eq. (5)

where the difference between quadrupolar and magnetic scattering enters. Therefore the enhancement of T_c is moderate despite the large change in λ.

Figure 3(b) shows the calculated frequency dependence of the real part of the superconducting gap and the renormalization function for $PrOs_4Sb_{12}$. One notices a sharp feature at an energy of about $\Delta_{CEF} + \Delta_0$ and a strong renormalization of the quasiparticle mass due to the presence of the low-energy exciton. It is interesting to note that both magnetic and quadrupolar scatterings contribute to the renormalization of the quasiparticle mass while only their difference contributes to Cooper-pair formation. We suggest that the low-energy peak in $\Delta(\omega)$ should be visible in the tunneling density of states which could be checked experimentally. Another remarkable feature is that we find $2\Delta_0/k_BT_c \approx$5.4 typical for a strong-coupling superconductor. Therefore, within this approach we find $PrOs_4Sb_{12}$ to be a conventional *s*-wave strong-coupling superconductor, with enhanced T_c due to low-energy excitons with mainly quadrupolar scattering. It has been proposed previously that quadrupolar scattering may alone yield unconventional superconductivity in $PrOs_4Sb_{12}$ due to the exciton dispersion.[7,8] Indeed such a possibility cannot be excluded from our calculation. At the same time we find it difficult to reconcile with the experimental evolution of T_c in $La_{1-x}Pr_xOs_4Sb_{12}$ as a function x. In particular, assuming the linear dependence of the exciton's contribution to $H_\pm(\omega)$ as a function of Pr concentration we find good agreement between calculated and measured $T_c(x)$ [see Fig. 4(a)]. At the same time, unconventional superconductivity in $PrOs_4Sb_{12}$ would be unstable with respect to nonmagnetic La impurities and T_c should vanish at a small concentration of La. In Fig. 4 (b) we show the evolution of the effective mass as a function of Pr concentration. We find that the effective mass increases by a factor of 2.5 from $LaOs_4Sb_{12}$ to $PrOs_4Sb_{12}$ which agrees with dHvA data.[5] An inclusion of the higher-lying CEF energy levels provides stronger renormalization of the effective mass.[23] It is interesting to note that experimentally $\Delta C/T_c$ shows a nonlinear dependence on the La concentration, in particular close to $PrOs_4Sb_{12}$[17] which cannot be explained by our theory. Whether this is a sign of the first-order phase transition between different symmetries of the superconducting order parameter or is a result of the Pr-Pr interaction neglected in our theory remains to be understood both theoretically and experimentally. Finally, let us note that our theory allows us also to explain qualitatively the behavior of T_c in $Pr(Os_{1-x}Ru_x)_4Sb_{12}$ for $x < 0.6$. In particular, it has been found that the CEF level splitting between the lowest levels increases as a function of x while T_c decreases.[24] According to our Fig. 1(a) the Cooper-pair constructive quadrupolar scattering decreases with increasing x while the magnetic pair-breaking contribution tends to saturate. Therefore, the T_c should decrease as it is also observed in the experiment.[24,25]

In conclusion by solving the nonlinear Eliashberg equations we find that in $La_{1-x}Pr_xOs_4Sb_{12}$ the dominant quadrupolar scattering due to the specific T_h crystal group symmetry of the lattice is responsible for the observed increase of T_c as a function of Pr concentration. Our analysis suggests that a combination of conventional electron-phonon interaction together with pairing mediated by quadrupolar excitons yields strong-coupling superconductivity in the $PrOs_4Sb_{12}$ system.

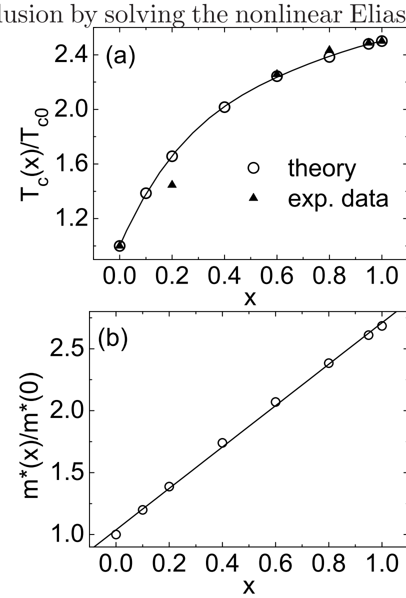

FIG. 4: Calculated superconducting transition temperature T_c (a) and quasiparticle effective mass m^*(b) for $La_{1-x}Pr_xOs_4Sb_{12}$ as a function of Pr concentration x. The straight curve is a guide to the eye. The experimental data are taken from Ref. 12.

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